

## Modeling the Multiphase Atmospheric Chemistry of Launch Clouds

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Prepared by

B. B. BRADY and L. R. MARTIN  
Propulsion Science and Experimental Mechanics Department  
Space Materials Laboratory  
Laboratory Operations

Prepared for

SPACE AND MISSILE SYSTEMS CENTER  
AIR FORCE MATERIEL COMMAND  
2430 E. El Segundo Boulevard  
Los Angeles Air Force Base, CA 90245

Space Systems Group

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# Modeling the Multiphase Atmospheric Chemistry of Launch Clouds

B. B. Brady\* and L. R. Martin†

The Aerospace Corporation, Los Angeles, California 90009-2957

The adaptation is described of a widely available subroutine library, originally developed to model chemical vapor deposition to model a plume of steam and hydrogen chloride gas that is released during solid rocket booster launches such as the Space Shuttle and the Titan IV. Hydrogen chloride is a very hygroscopic gas, and it tends to form an aerosol cloud from the water vapor in the atmosphere as well as from the steam released by engine combustion and launch deluge water. This aerosol cloud is the most difficult feature to model because it involves phase changes, complex thermodynamic data for HCl/water solutions, and rapid changes in temperature, concentration, and relative humidity. The model predictions are compared to field data taken during a launch of the Space Shuttle.

## Nomenclature

$A$	= preexponential factor from rate expression, $\text{cm}^3/\text{mol-s}$
$b$	= temperature exponent in rate expression
$E$	= activation energy in rate expression, $\text{cal/mole}$
$k$	= rate constant $AT^b e^{-(E/RT)}$ , $\text{cm}^3/\text{mol-s}$
$R$	= gas constant
$r$	= aerosol radius
$T$	= temperature, K
$V_L/V_g$	= liquid volume/gas phase volume in plume
$x$	= mole fraction
$\gamma$	= activity

## Introduction

**T**OXIC gases such as HCl,  $\text{NO}_2$ , or hydrazine fuels are released into the atmosphere as engine exhaust products, as a result of inadvertent spills of propellants or due to a launch abort. Monitoring the atmospheric concentrations of such gases and modeling their transport and transformation are essential to the evaluation of their impacts on the environment. Such impacts are an important consideration in launch constraints. The purpose of this report is to document a modeling process with higher accuracy, to convey the results of this modeling, and to discuss the implications for space launches.

Monitoring toxic gases in the field is complicated by the fact that HCl and  $\text{HNO}_3$ , a major conversion product of  $\text{NO}_2$  in the troposphere, are very hydrophilic and will tend to condense on atmospheric aerosols. Once such gases become incorporated in the aerosols, they will not appear on gas phase sensors unless provision is made to evaporate the aerosols in the analysis scheme. Furthermore, the environmental fate of such gases may be governed by chemistry in the aerosol and the motion of the aerosol in the atmosphere, rather than by gas phase chemistry alone. The presence of the aerosol will also affect the buoyancy of the pollutant cloud through both thermal and density effects.

The predicted path and exposure level of toxic gas from launches and launch abort scenarios are a factor in determining launch windows. At present, the models used for this purpose, such as the rocket exhaust effluent diffusion model (REEDM) code and the U.S. Air Force toxic chemical dispersion model (AFTOX), do not realistically deal with chemical transformation in the atmosphere. REEDM and AFTOX include chemistry via the source term; once the plume begins to disperse, its chemical composition is fixed. The latest version of REEDM does calculate the equilibrium concentrations from

an abort and uses them as a source term. As a result, these models can significantly overstate the hazard at a given distance downwind of the spill because atmospheric chemical transformation may reduce the noxious gas concentration. Thus, the launch window may increase as a result of more rigorous calculations.

The current models include a first-order loss of the toxic material to account for chemical transformation. These corrections are not based on realistic physics or chemistry and are not likely to be very reliable.

We have developed submodels that can deal with chemical transformation in a realistic way. We have converted the heterogeneous chemical kinetic model SURFACE CHEMKIN<sup>1</sup> into an atmospheric model that can deal with all of the homogeneous and heterogeneous chemistry encountered in a toxic launch cloud. These models have been applied to the case of a nitrogen tetroxide spill<sup>2</sup> and the HCl from solid rocket motor exhaust in the stratosphere.<sup>3</sup> Here we present the results from applying the same models to the HCl generated by the solid rocket boosters in a normal launch at ground level. The results of these new models need to be incorporated into dispersion models such as REEDM to better forecast the consequences of normal or aborted launch. If downwind chemical composition can be calculated in this model, it can be used as a source term for a model such as REEDM or AFTOX with little loss in accuracy.

## Model

We have developed a realistic kinetic model for multiphase atmospheric chemistry based on the SURFACE CHEMKIN model from Sandia National Laboratories. The purpose of the current calculations is to explore the effect of gas phase and aerosol chemistry on the downwind concentrations of HCl; if the concentrations are higher or lower than those predicted by pure dispersion models, a correction can be made to the current models, which will affect the toxic hazard corridor and influence launch availability. The details of the model have been described previously.<sup>2,3</sup> We modified the reaction manifolds in the current case to reflect the HCl chemistry. We have added the acid dissociation of the parent HCl into aqueous ions,  $\text{H}^+_{(\text{aq})}$  and  $\text{Cl}^-_{(\text{aq})}$ . Likewise, we have included the dissociation of sea salt and of water; there is some competition between these processes in the natural marine aerosol. The complete set of reactions is given in Table 1. In addition, we have added the thermodynamic data of HCl water solutions,<sup>4,5</sup> i.e., activity coefficients that give the correct experimental vapor pressures of water and HCl above the solutions. The activity of each component is very nonlinear in the mixture:

$$\gamma_{\text{HCl}} = 44x - 350x^2 + 2.7e4x^3 - 2.5e5x^4 + 1.7e6x^5$$

$$\gamma_{\text{H}_2\text{O}} = 28 - 92x + 83x^2 - 2.5e - 3x^3 - 18x^4$$

where  $x$  is the mole fraction of the relevant species. We tested these fits to the activity coefficients by trying to reproduce the experimental data of Rhein.<sup>6</sup> Rhein's investigations determined whether

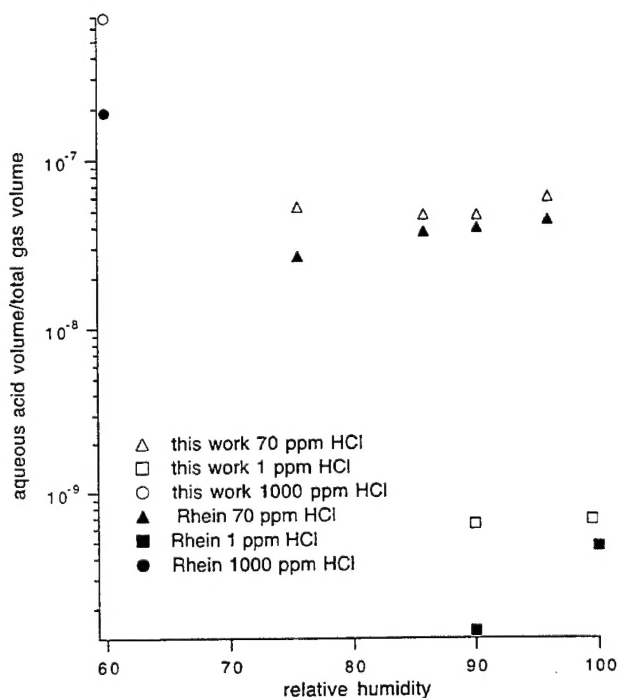
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\*Senior Member of the Technical Staff, Mail Stop M5-754, P.O. Box 92957.

†Senior Scientist, Mail Stop M5-754, P.O. Box 92957.

Table 1 Reactions considered in chemical model of launch cloud

No.	Reaction	A	b	E
<i>Gas phase reactions</i>				
1	$\text{OH} + \text{HCl} = \text{H}_2\text{O} + \text{Cl}$	$2.40\text{E}-12$	0	656.0
2	$2\text{Cl} + \text{M} = \text{Cl}_2 + \text{M}$	$6.14\text{E}-34$	0	-1,800.0
3	$\text{O} + \text{O} + \text{M} = \text{O}_2 + \text{M}$	$5.20\text{E}-35$	0	-1,790.0
4	$\text{O} + \text{NO}_2 = \text{NO} + \text{O}_2$	$6.50\text{E}-12$	0	-238.0
5	$\text{NO} + \text{O} = \text{N} + \text{O}_2$	$3.50\text{E}-15$	1.0	38,747.0
6	$\text{O} + \text{N}_2 = \text{NO} + \text{N}$	$1.30\text{E}-10$	0	75,506.0
7	$\text{N}_2\text{O} + \text{M} = \text{N}_2 + \text{O} + \text{M}$	$1.20\text{E}-06$	-0.7	62,789.0
8	$\text{O} + \text{N}_2\text{O} = \text{N}_2 + \text{O}_2$	$1.70\text{E}-10$	0	28,017.0
9	$\text{N} + \text{NO}_2 = \text{N}_2\text{O} + \text{O}$	$3.00\text{E}-12$	0	0
10	$\text{NO} + \text{M} = \text{N} + \text{O} + \text{M}$	$2.40\text{E}-09$	0	148,429.0
11	$\text{N}_2 + \text{M} = \text{N} + \text{N} + \text{M}$	$3.82\text{E}+05$	-3.5	225,008.0
<i>Surface reactions</i>				
12	$\text{H}_2\text{O} + \text{NaCl}_{(\text{sf})} \rightleftharpoons \text{NaCl}_{(\text{s})} + \text{H}_2\text{O}_{(\text{sf})}$ Coefficients are sticking parameters	$1.00\text{E}+00$	0	0
13	$\text{H}_2\text{O} + \text{H}_2\text{O}_{(\text{sf})} \rightleftharpoons \text{H}_2\text{O}_{(\text{sf})} + \text{H}_2\text{O}_{(\text{L})}$ Coefficients are sticking parameters	$1.00\text{E}+00$	0	0
14	$\text{HCl} + \text{H}_2\text{O}_{(\text{sf})} \rightleftharpoons \text{HCl}_{(\text{sf})} + \text{H}_2\text{O}_{(\text{L})}$ Coefficients are sticking parameters	$1.00\text{E}+00$	0	0
15	$\text{HCl}_{(\text{sf})} + \text{H}_2\text{O}_{(\text{sf})} \rightleftharpoons \text{H}^+_{(\text{sf})} + \text{Cl}^-_{(\text{sf})} + \text{H}_2\text{O}_{(\text{L})}$	$1.00\text{E}+10$	0	0
<i>Bulk reactions</i>				
16	$\text{NaCl}_{(\text{s})} + \text{H}_2\text{O}_{(\text{L})} \rightleftharpoons \text{Na}^+_{(\text{aq})} + \text{Cl}^-_{(\text{aq})} + \text{H}_2\text{O}_{(\text{L})}$	$1.00\text{E}+10$	0	0
17	$\text{H}_2\text{O}_{(\text{L})} + \text{H}_2\text{O}_{(\text{L})} \rightleftharpoons \text{H}^+_{(\text{aq})} + \text{Cl}^-_{(\text{aq})} + \text{H}_2\text{O}_{(\text{L})}$	$1.00\text{E}+10$	0	0
18	$\text{H}^+_{(\text{aq})} + \text{Cl}^-_{(\text{aq})} \rightleftharpoons \text{HCl}_{(\text{aq})}$	$1.00\text{E}+10$	0	0
<i>Transport</i>				
19	$\text{HCl}_{(\text{sf})} + \text{H}_2\text{O}_{(\text{L})} \rightleftharpoons \text{HCl}_{(\text{aq})} + \text{H}_2\text{O}_{(\text{sf})}$	$1.00\text{E}+10$	0	0

Fig. 1 Comparison of model predictions for aqueous acid volume fractions to data of Rhein.<sup>6</sup>

a given concentration of HCl in the gas phase at a given humidity and temperature will form an aerosol, the amount of aerosol, and the composition. We picked nine cases from Rhein's data covering the humidity range and HCl concentrations of the launch cloud. The model results are plotted in Fig. 1 along with Rhein's data; the aqueous acid volumes as a fraction of the total gas phase volume are plotted vs relative humidity for three different initial gas phase HCl concentrations. The model results are very sensitive to the activity.

Four phases are used to describe the reacting system in the model: the gas phase, the surface phase, an aqueous aerosol phase, and a solid aerosol phase. The solid aerosol phase consists of the alumina particles only; this phase participates in no reactions in the present model except to provide surface sites for condensation. The aqueous

aerosol phase consists of water, dissolved aqueous ions, hydrochloric acid, and sodium chloride. The chemistry is not limited by the rate of transport between phases,<sup>2</sup> and so we took the transport rates to be fast for simplicity.

The model is formulated as a two-bin system consisting of a plume bin that slowly mixes with the surrounding atmosphere and an ambient atmosphere bin that is not perturbed by the plume. The dispersion of the plume is based on a Gaussian plume model, Pasquill stability class C. The dispersion routine is a module of the program; other functions can be used for different meteorological conditions. The purpose of the current calculations is to test the impact of atmospheric chemistry on dispersion of toxic compounds; models now in use at launch sites include more sophisticated dispersion routines but make no allowance for chemistry.

The initial composition of the plume bin is determined in two ways. For the case of a light deluge or no deluge water, the output of the JANNAF plume model is used with some additional liquid water to simulate the light deluge condition on the launch pad. The deluge is a large flood of water across the launch pad designed to cool the pad and prevent the buildup of toxic exhaust products. The deluge begins several seconds before ignition of the first stage or booster stage and lasts until after the rocket has cleared the pad. Some small launch vehicles do not use a deluge. For the case of the actual Titan IV or Shuttle deluge, the direct output of the nozzle is used with only enough additional air to complete the combustion: roughly two volumes of air per volume of exhaust gas. In the JANNAF plume model starting conditions, 10 times as much air is used; the larger volume of air is equivalent to waiting 2 s after the exhaust leaves the nozzle before beginning our calculation. The large volume of water used in the Titan IV deluge system is a major perturbation on the plume, cooling it to ambient temperature within a few seconds.

### Description of Results

Several different initial conditions were investigated: three ambient relative humidities, 30, 60, and 90% at 293 K; and three deluge water amounts, 1) maximum deluge, enough to cool the plume to ambient temperature, equivalent to a Titan IV or a Space Shuttle deluge, 2) intermediate deluge, 1.5% of the maximum value, and 3) no deluge water, such as a Peacekeeper missile. The results always show the formation of a liquid aerosol that contains 7–93% of the HCl from the exhaust. In the maximum deluge case, the ambient humidity makes little difference, as expected. At the lowest humidity

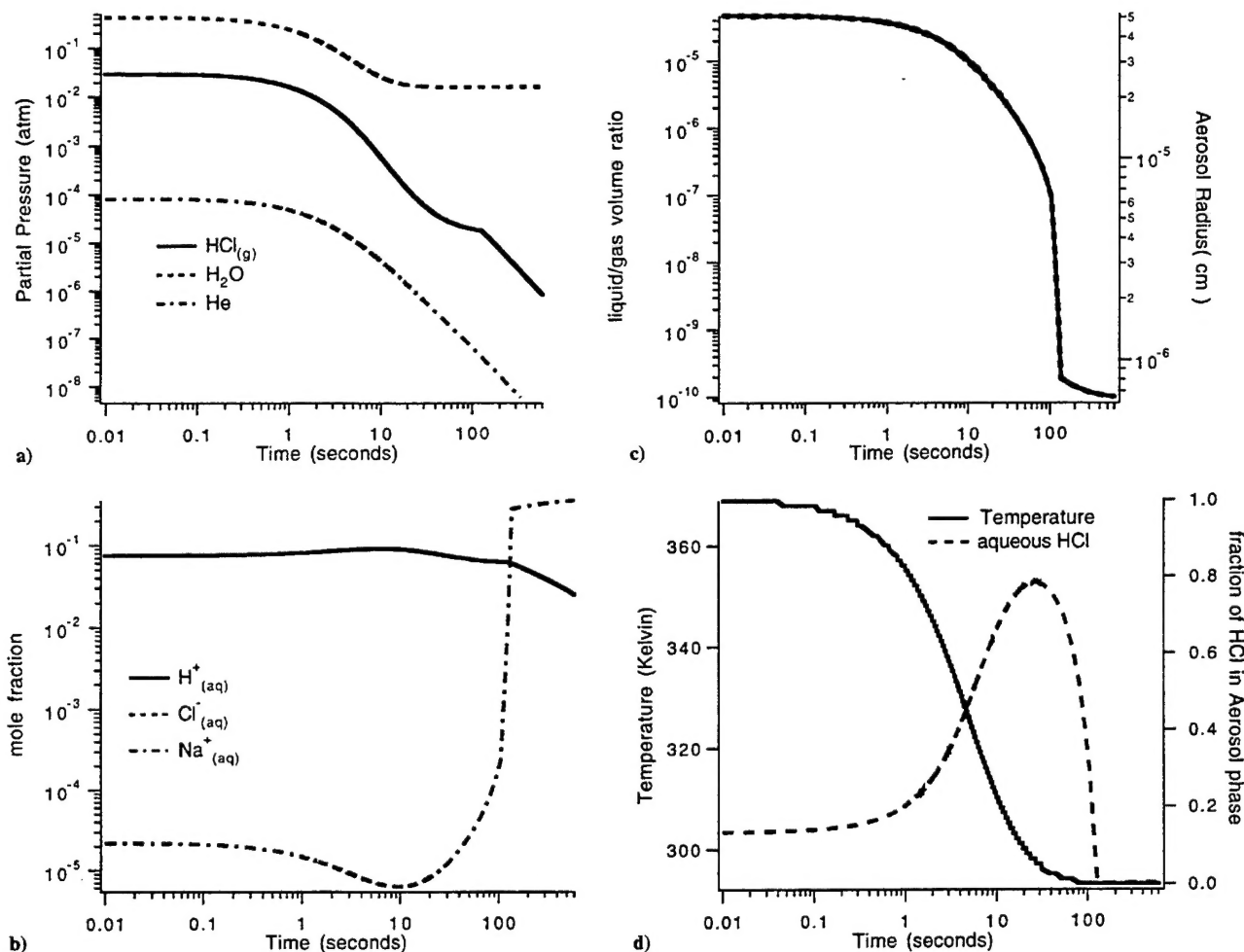


Fig. 2 Time evolution of a parcel of the plume with heavy deluge, 60% relative humidity.

in the maximum deluge case, the overall fraction of HCl is 58% in the aqueous phase and 42% in the gas phase.

Model results are shown in Fig. 2 as the log of the species concentrations ( $\text{HCl}$ ,  $\text{H}_2\text{O}$ , He,  $\text{H}^+_{(aq)}$ ,  $\text{Na}^+_{(aq)}$ , and  $\text{Cl}^-_{(aq)}$ ) vs log of the time, as well as  $r$ ,  $V_t/V_g$ , and temperature vs log of the time for the case of a normal Titan IV deluge at 60% relative humidity. Figure 2 shows the development of the groundcloud from a Titan IV launch with a heavy deluge and relative ambient humidity of 60%. The water vapor content of the cloud starts off at a high level because the deluge water is turned into steam. The water vapor content drops as the cloud expands and cools until it is the same as the surrounding air, and then it levels off. Helium is included in our plume at 100 parts per million by volume (ppm) as an artificial tracer; the helium shows how the concentration of a nonreactive gas changes as the plume dilutes. The helium concentration is flat at early times and then decreases as the square of time, the same rate at which the area of the plume is expanding. The concentration of hydrochloric acid gas ( $\text{HCl}$ ) in the plume decreases faster than the helium concentration because the  $\text{HCl}$  is dissolving in the liquid aerosol as well as being diluted by the surrounding air. After 30 s the  $\text{HCl}$  concentration flattens out; at this point the aqueous  $\text{HCl}$  serves as a reservoir, replenishing the gaseous  $\text{HCl}$  as it is diluted. At 100 s, the reservoir of aqueous  $\text{HCl}$  is depleted, and the gas phase  $\text{HCl}$  concentration begins to drop again at the same rate as the helium. The aqueous ion concentrations are shown in Fig. 2b. It is obvious that  $\text{HCl}$  ( $\text{H}^+/\text{Cl}^-$ ) is the dominant dissolved species in the liquid aerosol. The aerosol pH is between one and two for the entire simulation,  $0.1 > [\text{H}^+] > 0.01$  mole fraction. Figure 2c shows the aerosol volume (relative to the gas phase volume) and aerosol particle radius. In our simulation the number of aerosol particles is fixed; therefore, the volume and the radius are proportional and the curves overlap. The aerosol is initially at a volume fraction of 50 ppm and

a mean particle radius of  $0.5 \mu\text{m}$ . The solid alumina in the exhaust initially accounts for 1 ppm volume fraction and a mean particle size of  $0.14 \mu\text{m}$ . The ambient air contains a natural marine aerosol, 0.2 parts per billion by volume (ppb), which is composed of sea salt and water. Figure 2d shows the temperature, which is close to 380 K initially when the aerosol is at its peak size. The cloud cools as ambient air mixes in and the aerosol evaporates. Figure 2d also shows the fraction of  $\text{HCl}$  that is in the aqueous phase. The aqueous  $\text{HCl}$  is calculated as the moles of  $\text{HCl}$  in the aerosol divided by the sum of the gas phase moles of  $\text{HCl}$  and the moles of  $\text{HCl}$  in the aerosol. The fraction of aqueous acid peaks at 79% at 30 s and then drops rapidly. This curve is consistent with Fig. 2a, which shows the gas phase  $\text{HCl}$  stabilizing at about 30 s.

The other initial conditions we explored yield plots similar to Fig. 2. The main differences are in the timescale and the peak amount of  $\text{HCl}$  in the aerosol phase. Figure 3 shows the peak fraction of aqueous  $\text{HCl}$  and the time at which the peak is reached vs relative humidity for the three deluge cases. At higher relative humidities, the reactions proceed longer, and more  $\text{HCl}$  is absorbed into the aerosol droplets. At the highest humidity, 90%, the deluge water has little impact on the fraction of aqueous  $\text{HCl}$ , although it does affect the timescale. At lower humidities the deluge water can substitute for some of the atmospheric water, causing a greater difference between the heavy and light deluge cases. At all humidities, there is little difference between the light and no deluge cases. A common feature of the light and no deluge cases is that the aerosol volume fraction reaches a peak slightly before the peak in aqueous  $\text{HCl}$ ; for the heavy deluge cases, the aerosol volume is at maximum initially and decreases monotonically with time as in Fig. 2.

The maximum radius of the monodisperse aerosol particles varies from 5.5 to  $1.0 \mu\text{m}$ . The deposition velocity of particles this size is between 0.3 and  $0.03 \text{ cm/s}$ , compared to 3–5 cm/s for the  $\text{HCl}$  gas.

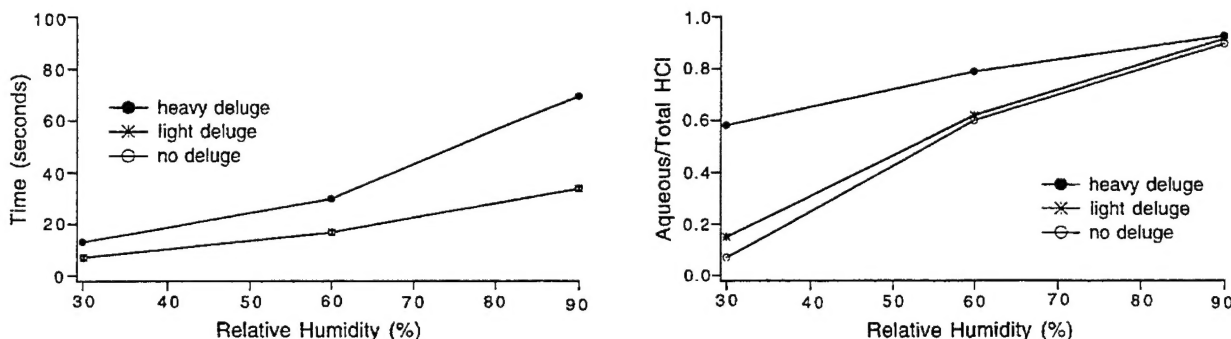


Fig. 3 Summary of runs with differing ambient relative humidity and deluge conditions.

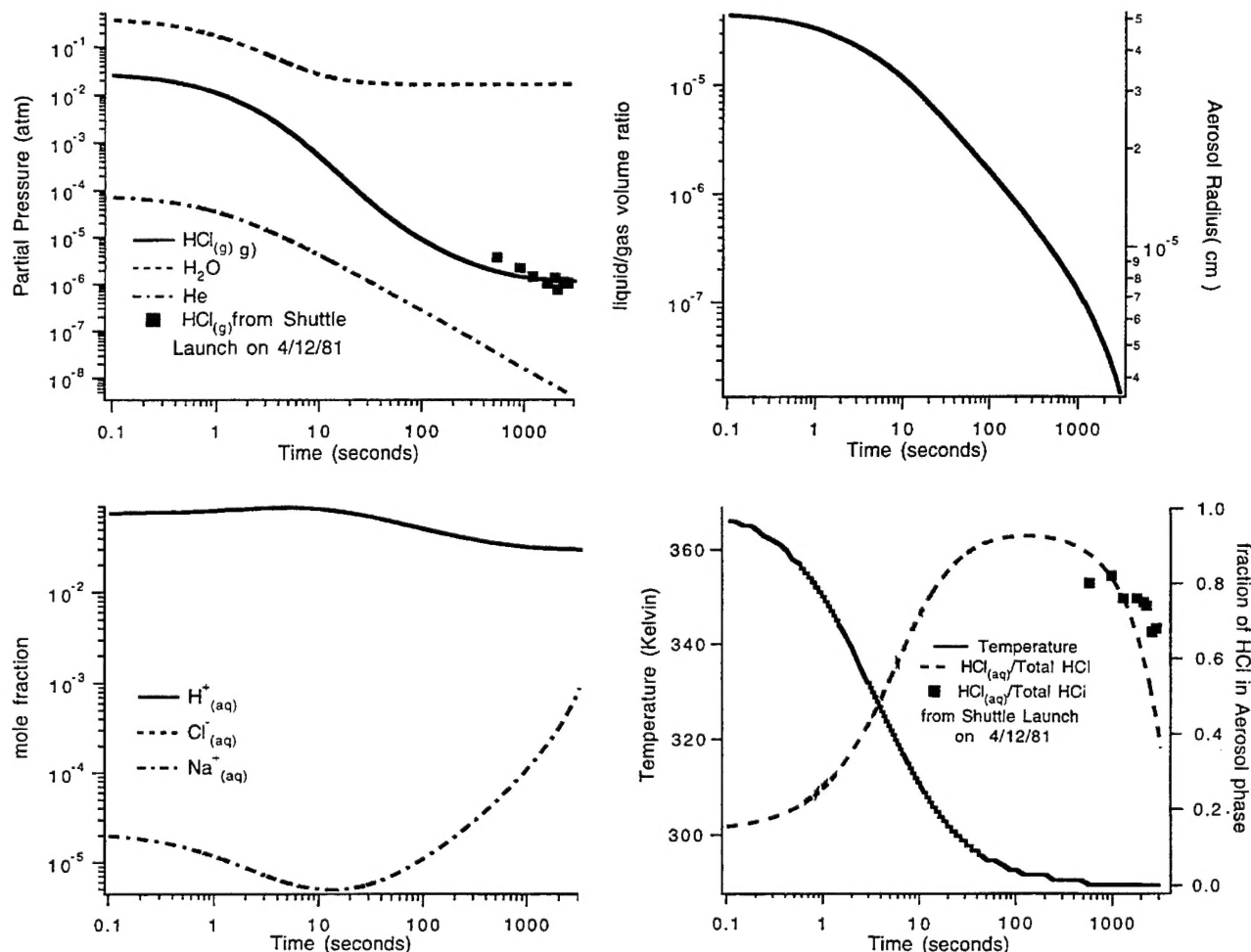


Fig. 4 Field data from the Shuttle launch on April 12, 1981 (■), compared to model output (see Fig. 2)

The aerosol lifetime depends steeply on the deluge water amount as well as the ambient humidity, varying from about 6 to 600 s. The aerosol evaporates in from 10 min to an hour depending on dispersion. The net result of the aerosol formation is to delay dry deposition of the exhaust HCl for 10 min to an hour.

The plume is also buoyant in each simulation. The presence of an aerosol and the HCl add to the density of the plume, but the water vapor and elevated temperature reduce the density. In all cases, the water vapor effect dominates, and the plume is always buoyant. This means the plume will rise, further limiting the advantageous effects of dry deposition. In a normal atmosphere, the plume will rise until it hits an inversion layer.

#### Comparison with Field Measurements

Data on the composition of launch clouds and plumes are scarce; however, a measurement of gaseous and total HCl was made after a

Shuttle launch in 1981 (Ref. 7). The Space Shuttle and the Titan IV both use solid rocket boosters of substantially the same composition. The Shuttle boosters are bigger, producing roughly 2 tons/s of HCl after ignition compared to 1 ton/s for the Titan IV. The Titan IV deluge system delivers 75,000 gal of water per minute while the engines are firing, which is 5 tons/s. The Space Shuttle deluge system delivers 8.3 tons/s of water. Combustion in the solid motor and afterburning generate about half a ton of water vapor per second for the Titan, twice as much for the Shuttle. In a Titan launch, only the solid boosters are firing, whereas in the Space Shuttle the main engine is also firing and it produces an additional 2 tons/s of water vapor. In each case, the mass ratio of water vapor to hydrogen chloride is about 5.5; this means we can use the Titan IV deluge conditions to model the Space Shuttle launch. The field measurements were made on two portions of the exhaust cloud, a high cloud that broke through the inversion layer and a low cloud that stayed at 650–950-m

altitude, below the inversion layer. The ambient conditions for the low cloud were a temperature of 290 K and 70% relative humidity. The dispersion rate used for the preceding examples (Figs. 2 and 3) were for Pasquill stability class C; these conditions are commonly observed at Vandenberg Air Force Base (VAFB). For the Shuttle launch, which is at Cape Canaveral, we fit the data on total HCl concentration vs time to the observed dispersion rate. Note that the dispersion rate observed at the Cape is much slower than the rate used for VAFB. In Fig. 4, the field measurement data on gaseous HCl and the fraction of HCl in the aerosol are plotted (solid squares) against the model predictions. The aerosol fraction was calculated from the difference between the total HCl and gas phase HCl divided by the total. The agreement between the model predictions and the field measurements is adequate.

### Conclusions

We have developed a realistic kinetic model for multiphase atmospheric chemistry based on the SURFACE CHEMKIN model from Sandia National Laboratories. Our calculations indicate that an HCl-water aerosol will form after any space launch using solid propellant. The fraction of HCl absorbed into the aerosol and the lifetime of the aerosol depend on the total amount of available water, as either deluge water, engine exhaust, or ambient humidity. In all cases, the aerosol evaporates and releases the absorbed HCl in from 10 min to an hour. The only effect the aerosol has on downwind concentrations then is to delay the onset of dry deposition because the aerosol droplets have a much smaller deposition velocity than the HCl molecule. This effect is expected to be minimal in contrast to previous results on other systems.<sup>2</sup> The plume is also buoyant in each simulation and in a normal atmosphere would be expected to

rise until hitting an inversion layer. The model has been partially verified by comparison to field data on gaseous and total HCl taken after a Space Shuttle launch.

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I. D. Boyd  
Associate Editor

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**Electronics and Photonics Laboratory:** Microelectronics, VLSI reliability, failure analysis, solid-state device physics, compound semiconductors, radiation effects, infrared and CCD detector devices, data storage and display technologies; lasers and electro-optics, solid state laser design, micro-optics, optical communications, and fiber optic sensors; atomic frequency standards, applied laser spectroscopy, laser chemistry, atmospheric propagation and beam control, LIDAR/LADAR remote sensing; solar cell and array testing and evaluation, battery electrochemistry, battery testing and evaluation.

**Space Materials Laboratory:** Evaluation and characterizations of new materials and processing techniques: metals, alloys, ceramics, polymers, thin films, and composites; development of advanced deposition processes; nondestructive evaluation, component failure analysis and reliability; structural mechanics, fracture mechanics, and stress corrosion; analysis and evaluation of materials at cryogenic and elevated temperatures; launch vehicle fluid mechanics, heat transfer and flight dynamics; aerothermodynamics; chemical and electric propulsion; environmental chemistry; combustion processes; space environment effects on materials, hardening and vulnerability assessment; contamination, thermal and structural control; lubrication and surface phenomena.

**Space Science Applications Laboratory:** Magnetospheric, auroral and cosmic ray physics, wave-particle interactions, magnetospheric plasma waves; atmospheric and ionospheric physics, density and composition of the upper atmosphere, remote sensing using atmospheric radiation; solar physics, infrared astronomy, infrared signature analysis; infrared surveillance, imaging, remote sensing, and hyperspectral imaging; effects of solar activity, magnetic storms and nuclear explosions on the Earth's atmosphere, ionosphere and magnetosphere; effects of electromagnetic and particulate radiations on space systems; space instrumentation, design fabrication and test; environmental chemistry, trace detection; atmospheric chemical reactions, atmospheric optics, light scattering, state-specific chemical reactions and radiative signatures of missile plumes.

**Center for Microtechnology:** Microelectromechanical systems (MEMS) for space applications; assessment of microtechnology space applications; laser micromachining; laser-surface physical and chemical interactions; micropropulsion; micro- and nanosatellite mission analysis; intelligent microinstruments for monitoring space and launch system environments.

**Office of Spectral Applications:** Multispectral and hyperspectral sensor development; data analysis and algorithm development; applications of multispectral and hyperspectral imagery to defense, civil space, commercial, and environmental missions.